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# MEASUREMENTS OF NATURAL RADIOACTIVITY IN THE SALT CAVERN OF THE POLKOWICE–SIEROSZOWICE COPPER MINE\*

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Due to their low radioactivity background, underground physics laboratories offer a unique possibility for investigating extremely rare phenomena like proton decay, dark matter signals or neutrino physics/astrophysics related issues. The knowledge of the natural radioactivity background is essential for the success of an underground physics experiment. The following measurements of the natural radioactivity background, in the foreseen location of an underground physics laboratory in the salt layer, in the Polkowice–Sieroszowice copper mine are presented: concentration of natural radio-isotopes from in situ obtained gamma-ray spectra and from alpha spectroscopy of rock samples, radon concentration in the air and the dose determination.

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## 1. Introduction

The depth, available surface/space for experiments, type of access (mine or tunnel) and natural radioactivity background are the main characteristics which differentiate underground physics laboratories. However, only two of them, namely the depth and natural radioactivity, have a direct influence on the sensitivity of the measurement of very rare events. The depth, *i.e.* shielding with rock overburden, may reduce the cosmic-ray muon flux by many orders of magnitude. Typically, at the depth of 1000 m (about

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2 700 m.w.e.) the muon flux is about 1–2 muons/m<sup>2</sup>/h, whereas at the surface (sea level) is about  $7 \times 10^5$  muons/m<sup>2</sup>/h. However, high energy neutrons generated by residual underground cosmic-ray muon interactions give complicated contribution to the background. Another sources of “underground” neutron background are ( $\alpha$ ,  $n$ ) and fission type reactions, originating from U and Th contamination in the rocks. The alpha decays of <sup>226</sup>Ra, which is present in the the rock and in the water, into gaseous radon <sup>222</sup>Rn, as well as alpha decays of other short lived members of U or Th series, contribute to the background, too. The ventilation of the underground laboratory can significantly decrease the radon concentration in the air.

In this paper, we present the results of: (1) concentration of radioactive isotopes in the rock, from both *in-situ* and laboratory measurements, (2) radon concentration in the air and (3) the dose. The measurements have been performed in the P1 salt cavern in the Polkowice–Sieroszowice copper mine, belonging to the KGHM Polska Miedź S.A. holding. The salt layer with the cavern P1 has a thickness of about 70 m and is placed 930 m below the surface. The salt layer is taken into account as a possible location of an underground physics laboratory. That was the direct motivation to perform measurements there. A more detailed description of the Polkowice–Sieroszowice mine can be found in [1].

## 2. Natural radioactivity measurements

The set of measurements of natural radioactivity, exploiting different techniques, performed in the salt cavern P1 in the Polkowice–Sieroszowice copper mine is presented in this chapter.

### 2.1. *In-situ* measurements

The radioactivity background from natural radionuclides was measured (March 2005) by the *in situ* method using a portable gamma-ray spectrometry workstation. The M-1 *in situ* Ortec Industries system consists of a HPGe detector (30% efficiency, crystal length of 59 mm and diameter of 58.6 mm) with cryostat mounted on a tripod, a multichannel buffer DART and a laptop. The energy resolutions of the detector are 0.66 keV at 122 keV, and 1.73 keV at 1.33 MeV. The M-1 is a in-field soil analysis system. The “1-meter” methodology was developed by the U.S. Department of Energy Environmental Measurements Laboratory. It reduces the complex measurement problem to the product of three simply determined factors: efficiency calibration, attenuation correction and angular flux correction [2]. The radionuclide-specific spectral data are collected with a germanium detector 1 meter above the surface. The gamma-ray peak areas are related to a specific nuclide activity by the product of these three factors, which are

determined for a broad range of detectors and soil conditions. They are tabulated within the M-1 software. The spectrum of detected gamma-ray peaks, which are measured in the range 40–2700 keV, is presented in Fig. 1. The following concentrations of natural isotopes have been determined:  $^{226}\text{Ra}$  (from  $^{238}\text{U}$  series) equal to  $3.1 \pm 0.3 \text{ Bq/kg}$  (average from  $^{214}\text{Pb}$  and  $^{214}\text{Bi}$  peaks),  $^{232}\text{Th}$  equal to  $0.7 \pm 0.2 \text{ Bq/kg}$  (average from  $^{212}\text{Pb}$  and  $^{228}\text{Ac}$  peaks) and  $^{40}\text{K}$  equal to  $9.2 \pm 1.7 \text{ Bq/kg}$ . The integrated background counting rates in the energy range 40–2700 KeV is equal to  $2.30 \pm 0.02 \text{ CPS/keV/kg}$  (CPS means counts per second).

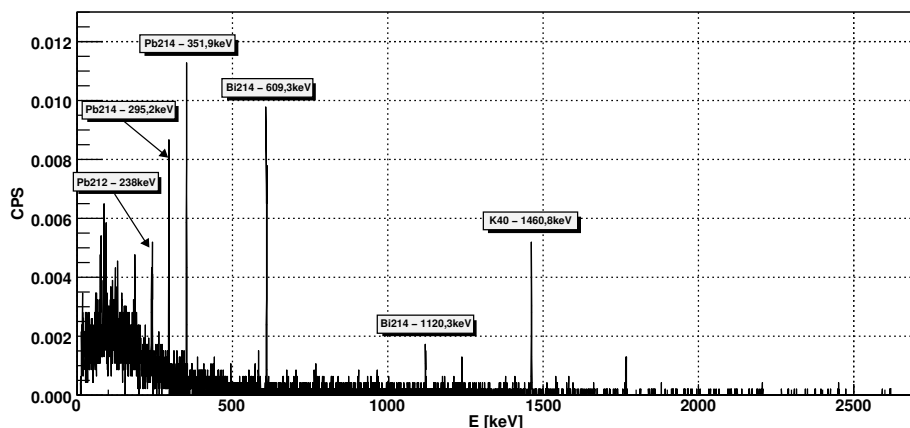


Fig. 1. The gamma spectrum measured in the salt cavern P1. Several, most pronounced peaks are indicated.

## 2.2. Measurements of the radon concentration in the air

The preliminary measurements of radon concentration in air were performed in the salt cavern at the depth of 930 meters under surface using AlphaGUARD PQ2000 ionization chambers (Genitron, Germany). Radon concentration was measured in five points: three of them were situated near cavern entrance, one in the middle, and one near the wall opposite to the entrance.

The AlphaGUARD is a reference device for radon measurements based on alpha spectroscopy (flow-in ionization chamber). It enables continuous monitoring of radon concentration and weather conditions at the site of the measurement. The PRO model is capable for operating in two alternative modes:

- diffusion mode with a 10- and 60-minute measuring cycle,
- flow mode with a 1- and 10-minute measuring cycle.

In the Polkowice–Sieroszowice mine the diffusion mode with 10-minute measuring cycle was applied in four points. The flow mode with 1-minute measuring cycle was applied in one point. Together with radon concentrations the weather conditions (air pressure, temperature and humidity) were recorded in each point. At points P2, P4 and P4a the photon dose rate was measured by AlphaGUARD PQ2000 PRO. The devices were placed 25 cm above surface. The measurements were performed during normal operation of mine (ventilation, car and machines traffic). The results are presented in Table I.

TABLE I

$^{222}\text{Rn}$  concentration measurements in the Polkowice–Sieroszowice mine’s air. (MP — measurement point, see text, AGM — AlphaGUARD PQ2000 PRO operating mode, see text,  $^{222}\text{Rn}$  —  $^{222}\text{Rn}$  concentration, DR — dose rate,  $T$  — air temperature, P — air pressure, H — air humidity.)

MP	AGM	$^{222}\text{Rn}$ [Bq m $^{-3}$ ]	DR [nSv h $^{-1}$ ]	$T$ [°C]	P [mbar]	H [%]
P1	Diff, 10 min.	$19 \pm 5(10, 38)$	—	33.6	1038	23
P2	Flow, 1 min.	$49 \pm 8(4, 101)$	9	24.2	1038	48
P3	Diff, 10 min.	$16 \pm 3(16, 16)$	—	35.0	1037	21
P4, 4a	Diff, 10 min.	$45 \pm 4(32, 52)$	3	34.9	1056	26
P5	Diff, 10 min.	$12 \pm 4(10, 13)$	—	35.3	1036	21

These results show that most of the radon in the salt cavern comes with the air introduced by ventilation.

### 2.3. Alpha spectroscopy measurements of rock samples

Four samples of salt rock from the Polkowice–Sieroszowice mine were analyzed for natural radioactivity in 2004 [3]. Measurements were done by means of low background gamma spectrometry with muon-veto shield and by alpha spectrometry (Silena Alphaquattro spectrometer) preceded by radiochemical procedure.  $^{232}\text{U}$  (equilibrated with  $^{228}\text{Th}$ ) as internal standard was added first. Since  $^{228}\text{Th}$  was also expected to be present in samples an additional check of radioactive equilibrium in Th series (between  $^{232}\text{Th}$  and  $^{228}\text{Th}$ ) was performed (a run without added tracer), which allowed to interpret the excess of  $^{228}\text{Th}$  above  $^{232}\text{Th}$  activity as yielding from added tracer. Blank sample was processed as well. The procedure consist of the following steps:

1. dissolution and complete mineralization in hot mineral acids;
2. pre-concentration of U and Th by  $\text{Fe}(\text{OH})_3$  co-precipitation at pH 9, it was necessary to remove majority of NaCl matrix from analyzed traces;
3. separation of U using anion-exchange resin Dowex  $1 \times 8$ : deposition 9 M HCl, elution: 0.5 M HCl with some reduction agent added ( $\sim 0.5$  g of hydroxyloamine hydrochloride);
4. separation of Th using anion-exchange resin Dowex  $1 \times 8$ , deposition 8 M  $\text{HNO}_3$ , elution: 10 M HCl;
5. preparation of alpha sources by  $\text{NdF}_3$  micro co-precipitation.

In 2006 another salt rock sample (originated from different part of salt deposit in the same cavern) and sample of anhydrite rock were analyzed. The  $^{229}\text{Th}$  tracer was available in laboratory, so it was added besides  $^{232}\text{U}$ . Salt sample was analyzed using similar procedure, whereas for powdered anhydrite the initial step was conversion of sulphates into carbonates in water bath, then complete dissolution in hot mineral acids. Next, the procedure followed steps 3–5 of procedure described above for salt samples. The radiochemical procedure consists of complete dissolution in hot mineral acids ( $\text{HF}$ ,  $\text{HNO}_3$ , HCl with  $\text{H}_3\text{BO}_3$ ) in presence of  $^{229}\text{Th}$  and  $^{232}\text{U}$  tracers followed by anion exchange separation and  $\text{NdF}_3$  source preparation (similar to steps 3–5 of above).

TABLE II

Results on activity concentration (in Bq/kg) in rock salt and anhydrite samples from the Polkowice–Sieroszowice mine (\* — gamma spectrometric measurement, n.d. — not determined).

Sample		Mass [g]	$^{238}\text{U}$	$^{234}\text{U}$	$^{230}\text{Th}$	$^{232}\text{Th}$	$^{235}\text{U}$	$^{40}\text{K}$
1	salt '04	43.09 $\pm 0.06$	0.40 $\pm 0.06$	0.38 $\pm 0.05$	0.29 $\pm 0.03$	0.09 $\pm 0.006$	0.015	n.d.
2	salt '04	41.64 $\pm 0.05$	0.34 $\pm 0.05$	0.33 $\pm 0.06$	0.34 $\pm 0.02$	0.08 $\pm 0.007$	0.015	n.d.
3	salt '04	40.55 $\pm 0.02$	0.10 $\pm 0.02$	0.14 $\pm 0.03$	0.10 $\pm 0.02$	0.03	< 0.005	n.d.
4	salt '04	100.3 $\pm 0.02$	0.14 $\pm 0.02$	0.14 $\pm 0.03$	0.19 $\pm 0.02$	0.11 $\pm 0.004$	0.008 $\pm 0.3$	2.1*
5	salt '06	100.5 $\pm 0.002$	0.017 $\pm 0.002$	0.021 $\pm 0.001$	< 0.015 $\pm 0.001$	0.008	< 0.002 $\pm 0.9$	4.0*
6	salt '06	101.2 $\pm 0.003$	0.016 $\pm 0.003$	0.025	< 0.015 $\pm 0.001$	0.008	< 0.00 2	n.d.
7	anhydr.	8.35 $\pm 0.10$	0.82 $\pm 0.09$	0.76 $\pm 0.24$	1.26 $\pm 0.15$	0.52	< 0.05	n.d.

The results are presented in Table II. All Polkowice–Sieroszowice salt rock samples show very low concentration of radioactive traces. They are within an order of magnitude of what can be a result of the processes which occurred during formation of deposit. Even the anhydrite rock has much lower radioactivity as compared to granite. The results combined with the depth of almost 1 km explain the observed very low external dose rate within the Polkowice–Sieroszowice mine.

In salt mine, where no water is present, it can be assumed that radionuclides within U and Th series are in equilibrium. However, for almost all salt samples  $^{234}\text{U}$  ( $T_{1/2} = 2.45 \times 10^5$  years) and  $^{230}\text{Th}$  ( $T_{1/2} = 7.57 \times 10^4$  years) seem to be not yet equilibrated which might suggest water inflow episodes in the past on scale of thousands of years.

#### *2.4. Dose measurements*

The kerma in air ( $K_{\text{air}}$ ) rate from external ionizing radiation in salt cavern was measured using high sensitive thermoluminescent MCP-N (LiF:Mg, Cu,P) detectors. Detectors were annealed in PTW TLDO (PTW, Freiburg) furnace and readout in manual TL reader in mine. All readout and annealing conditions were according to standards used by the Laboratory of Individual and Environmental Dosimetry at IFJ PAN. Due to the low background, detectors were exposed for 243 days (24.03.2005–22.11.2005). They were packed in dosimeter badges and sealed in the PTFE foil to avoid any contact with salt dust which is thermoluminescent. 78 MCP-N detectors were placed on the wall of salt cavern in many sites. The average dose rate was 1.8 nGy/h. For comparison typical background in Poland measured 1 m above ground is on the level of 65 nGy/h. Results obtained in the Polkowice–Sieroszowice mine agree well with those obtained during 1.5 year of measurements of natural background in Asse Salt mine near Salzgitter, Germany. Using the same type of high sensitive detectors the average dose rate was 1.1 nGy/h [4].

### **3. Conclusions**

The results of natural radioactivity measurements performed in the salt cavern P1 in the Polkowice–Sieroszowice mine have been presented. The values of Th and U concentrations are much smaller (less than 1 Bq/kg) than those for typical rock (about 20–30 Bq/kg). Such, extremely low concentrations support very strongly the Polkowice–Sieroszowice mine salt layer for the location of a low background underground laboratory.

Unlike in other underground locations (caves, coal mines, mining adits), the obtained radon concentrations are similar to radon levels observed in open air. The salt layer seems to be a good protection against radon exhalation from deeper geological structures. Radon presence is caused mainly by regularly working ventilation in this mine. As the next step radon measurements without ventilation are planned.

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